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# EFFECTS OF CHLORINE DIOXIDE AND SOME WATER QUALITY PARAMETERS ON THE FORMATION OF THMS IN WATER TREATMENT PLANTS

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## ABSTRACT

The presence of trihalomethanes (THMs) in drinking water has attracted the attention of both researchers and professionals, because of the harmful effects of these substances on human health. Chlorine dioxide ($ClO_2$) is an alternative to chlorine because it is an oxidizing agent rather than a chlorinating agent, and therefore, will not form chlorinated disinfection byproducts such as THMs under typical water treatment conditions.

The present study was conducted to determine the effects of Chlorine Dioxide, Temperature, Turbidity, Hydrogen ion concentration (pH), Total Organic Carbon (TOC), and contact time on the formation of THMs during treatment process. A small-scale pilot plant of a compact model is designed and constructed at Environmental Engineering Department lab (Babylon University).It is simple in operate and made of galvanized iron of its components and locally available materials.

Total Trihalomethanes (TTHMs), their speciation compounds (THM4), and varieties of water quality parameters were monitored by addition different doses of chlorine dioxide and alum to raw water pipe entering the pilot plant. The input flow rate was (0.36 $m^3$/hr) with characteristics (as average) (Temperature, Turbidity, Alkalinity, TTHMs, pH, and TOC) are (28 0C, 27 NTU, 140mg/L, 0.1545 mg/L, 8.22, 1.4 mg/L) respectively.

All runes were done during April, May and June of 2012. The linear correlations were calculated between THM concentrations and $ClO_2$ dose or selected water quality parameters, in an attempt to identify one or more parameters that could be used as surrogate in THMs investigation. (HS – GC - ECD) with Gas chromatography analysis techniques were used to measure the THMs concentrations. It was noticed that TTHMs concentration increases as temperature, TOC, and pH increase, and average TTHM levels detected in all runes, does not exceeded the USEPA's Stage I (limit of 80 µg/L), while exceeded the stage II limit in all sampling measurements.

**KEYWORDS:** Trihalomethane, Drinking Water, Temperature, Turbidity, pH, TOC

## INTRODUCTION

In **1976** the **U.S. National Cancer Institute** published a report linking chloroform to cancer in laboratory animals. Two years previously, **(Rook, 1974),** was the first to report on tile occurrence of this compound in chlorinated drinking water. During the ensuing years water with disinfectant chemical reagents, as a result of which a long and varied list of disinfection by-products (DBPs)--many of which might be teratogenic, mutagenic and/or carcinogenic-has been compiled (**Fawell and Hunt, 1988**) in which the trihalomethanes (THMs) are only the tip of the iceberg **(Klein, 1990).** The concern of the national and supranational health authorities has resulted either in recommendations or regulations about the maximum levels of several groups of DPBs **(EEC, 1980; USEPA, 1988; WHO, 1992**) and has prompted studies

on the reaction mechanisms and conditions involved and, furthermore, on developing kinetic computer models to simulate the formation of DBPs during water treatment, for both their control and minimization. The information on the reaction mechanism of the formation of THMs is still limited, although it is generally recognized that four factors would be involved in their formation: the chlorine-to-precursor molar ratio, pH, temperature and reaction time.

THM formation is strongly dependent upon the chlorine concentration (**Kavanough et al., 1980; Peters et al., 1980**). However, there is some disagreement regarding the quantitative relations between chlorine concentrations and the rate of THMs production, linear relationship between chlorine consumption and the production of THMs with a reaction order greater or equal to unity (**Kavanough et al., 1980; Trussel and Umphres, 1978**). Despite this, it is also possible that the reaction order might change during the course of the reaction (**Kavanough et al., 1980).**

The formation of THMs also increases strongly with increasing amounts of soluble organic matter, following a first-order reaction. In naturally occurring water, this organic matter usually consists of humic substances (**Trussel and Umphres, 1978; Babcock and Singer, 1979**). Although fulvic acid accounts for over 90% of the aqueous humics in many water sources, **Babcockand Singer** found that relative contributions to the formation of THMs by the humic fraction is greater than that of the fulvic fraction since the former substances react more readily with chlorine.

Increased pH values lead to increases in THMs formation (**Stevens et al., 1989; Onodera et al, 1987),** three-fold increases being reported in the reaction rate per unit of pH (**Kavanough et al., 1980).** The lower the pH, the higher non-ionized HClO form of hypochlorous acid is found, thus increasing its reaction rate with the humic matter. However, THM yields depend rather on the last step of the THM reaction pathway, which is base-catalyzed as with the haloform reaction (**Simmon and Tardiff, 1978).** These findings have also been reported by other authors (**Peters et al., 1980; Sandier, 1977**). According to **Abedin et al., (2010)**, the acidic functional groups of humic matter are not ionized, leading to the aggregation of molecules due to Van der Waals forces. This phenomenon is also associated with folding of the huraic molecules, leaving fewer sites available for attack by chlorine (**Trehy and Bieber, 1980),** thereby red'acing THM production. In studies on the effect of temperature on THMs formation, an Arrhenius-type dependence has been found between the rate constant and temperature, with activation energies ranging from 10-20 kJ mol (**Peters et al, 1980)** to below l0 kJ mol(**Kavanough et al., 1980; Stevens et al., 1989**). Accordingly, a higher rate of THM formation should be expected with increasing water temperatures although, on the other hand, the volatility of these compounds should account for their partial remotion in open systems.

## EXPERIMENTAL WORK

This paper deals with the analyses and discussion of the results of experimental work to show the effect of $ClO_2$, onthe formation of THMs and other water quality parameters. Total Trihalomethanes (TTHMs), their speciation compounds (THM4), and varieties of water quality parameters were monitored in asmall-scale pilot plant. By addition different doses of chlorine dioxide and alum to raw water pipe entering the pilot plant and monitoring THMs concentration. The raw and finished water quality parameters which were analyzed are included; Hydrogen ion concentration, Temperature, Total organic carbon, Turbidity, Total Trihalomethane concentration and examines the percent reduction of Trihalomethanes formation with applying different doses of chlorine dioxide. All runes were done during April, May and Juneof 2012. Thelinear correlations were calculated between THM concentrations and $ClO_2$ dose or selected water quality parameters, in an attempt to identify one or more parameters that could be used as surrogate in THMs investigation. The input flow rate was ($0.36 m^3/hr$) with characteristics listed in Table (1):

**Table 1: Characteristics of Raw Water**

| Parameter | | Value, (Average) |
|---|---|---|
| Temperature, ($^0$C) | | 28 |
| Turbidity, (NTU) | | 27 |
| Alkalinity, (mg/L) | | 140 |
| THMs (mg/L) | CF | 0.02400 |
| | BDCM | 0.05257 |
| | DCBM | 0.0588 |
| | BF | 0.01905 |
| TTHMs, (mg/L) | | 0.1545 |
| pH | | 8.22 |
| TOC, (mg/L) | | 1.4 |

## TTHM SPECIATION IN THE STUDY

In this study another point of interest was the THM speciation. Figure 9 presents a representative THM speciation. This figure summarizes the weight fractions of $CHCl_3$, $CHBrCl_2$, $CHBr_2Cl$, and $CHBr_3$ species. Results show that the intermediate brominated species ($CHBrCl_2$, and $CHBr_2Cl$) comprised most of the THM concentrations. However, results of recent toxicological studies suggest further attention should be paid to the occurrence of brominated compounds, especially bromodichloromethane ($CHBrCl_2$) **(Krasner et al., 1992)**.

In general, all results indicate that the treated water contain mainly the brominate THM species ($CHClBr_2$ and $CHBrCl_2$). The average weight fraction of chloroform ($CHCl_3$) compound ranged from 7.16% to 21.46%, while the average weight fraction of the rest brominated ($CHBr_2Cl$, and $CHBrCl_2$) species ranged from about 34.97% to 43.8%, 30.67% to 34.54% respectively as shown in Figure 1. Nearly the same pattern was pointed out by **(McLain et al., 2002).**

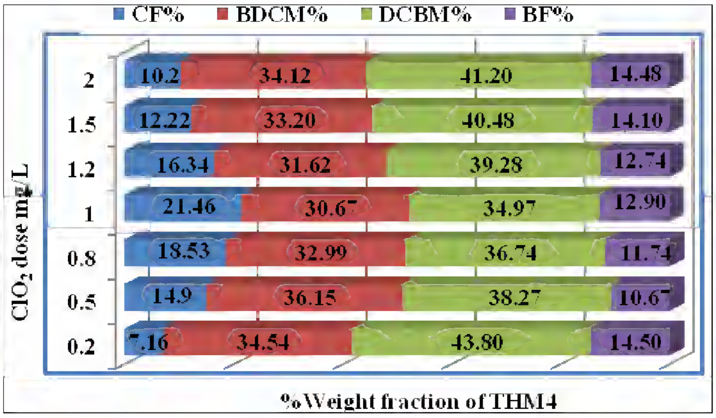


**Figure 1: Weight Fraction of Individual THM4 to Total THM4**

## EFFECTS OF CHLORINE DIOXIDE DOSE ON TRIHALOMETHAN FORMATION

The results showed that the THM formation decrease with increasing chlorine dioxide dose, concentrations of THMs that developed in water pretreated with 2 mg/L $ClO_2$were significantly lower when compared to those in water pretreated with the other dose of $ClO_2$.It is observed that higher THM level at $ClO_2$ dose of 0.2 mg/l and then tends to decrease slightly with increasing $ClO_2$ dose.Increasing$ClO_2$ doses have been shown to have the opposite effect as chlorine. This finding is agreed with the results adapted by **(Griese, 1991).**

Also the results showed that with increasing dose of $ClO_2$ from (1.0 to 2.0) mg/Lleads to decrease reduction in THMs concentration with about (24) percent (**Griese, 1991; Hulsey et al., 2000)** were also found the same results.They were found that increasing doses of $ClO_2$ leads to increasing reduction in THM formation and approximately (20.74) percent reduction in THM concentrations when 1.0 to 2 mg/L of $ClO_2$ was applied.

Figure 2 shows the variation of TTHMs with chlorine dioxide dose used in the study. As shown the TTHMs formation decreasing with increasing chlorine dioxide dose.This Figurewas performed on the $ClO_2$ samples to determine whether $ClO_2$ dose affected the final TTHM levels.

While Figure 3 represents the percent reduction in TTHMs concentration of filtered water after pre oxidation with $ClO_2$. Fromthis Figure we found that the removal efficiency increases with the increase chlorine dioxide dose, similar result was found by (**Hulsey et al., 2000)** who concluded that the removal efficiency of THMs from water increase by increasing chlorine dioxide dose.

As expected, correlations are negative between THM4 concentrations and chlorine dioxide dose. During studyperiod, the regression equation indicates that higher percent removal of TTHMs was (64.33%) when using (2mg/L) of $ClO_2$ dose. The present results showed that the higher dose of $ClO_2$ is considerably efficient for the removal of THMs from raw water. The performance of the $ClO_2$in reduction of THMs formation can be expressed in the form Y=7.637X+46.76 with coefficient of determination ($R^2$) equal to 0.902

Figure 3 shows that the THMs removal efficiency is highly dependent on chlorine dioxide dosage. It is evident that at higher dosages (more than 1.5 mg/L), the $ClO_2$ started to achieve higher removal percentages of THMs. This can be attributed to the fact that the chlorine dioxide has higher capacity to removal THMs.

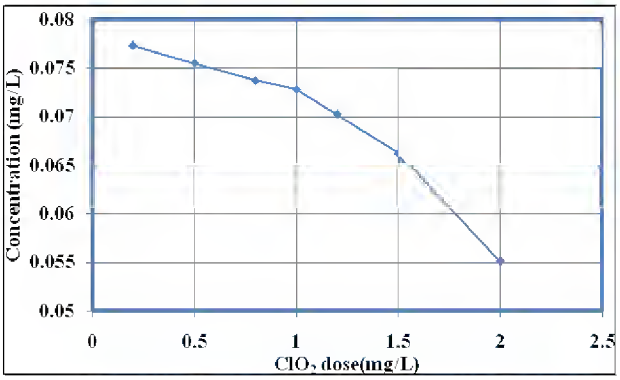


**Figure 2: The Variation of TTHMs with $ClO_2$ Dose**

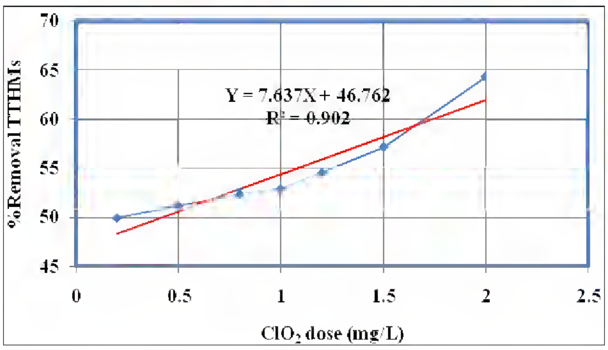


**Figure 3: The Percent Reductions in the TTHMs Concentrations after $ClO_2$ Dose**

Figure 4 presented a comparison of total THMs measurements with the USEPA Stage I and stage II limits. None of the THM levels exceeded the stage I limit, while, the stage II limit was exceeded in all sampling measurements.

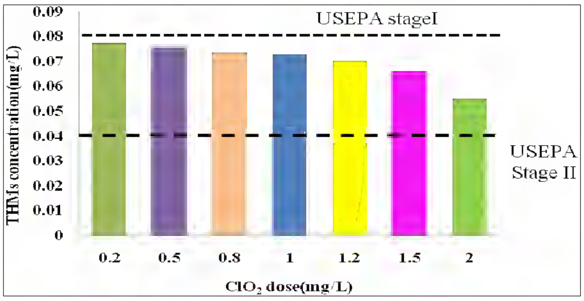


**Figure 4: Maximum THMs Levels Detected in the Study**

Figure 5 shows the variation of four THMs compound with $ClO_2$ dose.As shown the dominant TTHMs species during all runsis DCBM, it consists (39.2%) of trihalomethanes. It is decrease slightly with increasing dose, the remaining THMs are bromodichloromethan 33.34% which began slightly decrease with increasing dose, while the chloroform is 14.5%, and bromoformis 8.6%

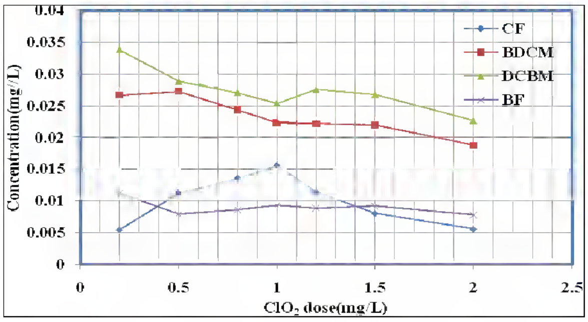


**Figure 5: Relative Amounts of THMs Compounds with $ClO_2$ Dose**

Figures 6 and 7 show the relation between chlorine dioxide dosing and percent reduction of two compounds (chloroform, and bromodichloromethane). Correlations are negative between chloroform percent reduction and chlorine dioxide dose under the dose (1mg/L), above this dose the percent began to increase, so the chloroform percent reduction increase with increasing chlorine dioxide dose above (1mg/L). Thus, we conclude that chloroform concentration is proportional with $ClO_2$ dose in the form (Y= 5.3961X+52.04) with coefficient of determination ($R^2$=0.0407). While correlations are positive between bromodichloromethane percent reduction and chlorine dioxide dose (decrease in BDCM concentrations with increasing chlorine dioxide dose). Thus, we conclude that BDCM concentration is proportional with $ClO_2$ dose in the form (Y= 8.846X+46.408) with coefficient of determination ($R^2$=0.9122) as shown in Figure 7.

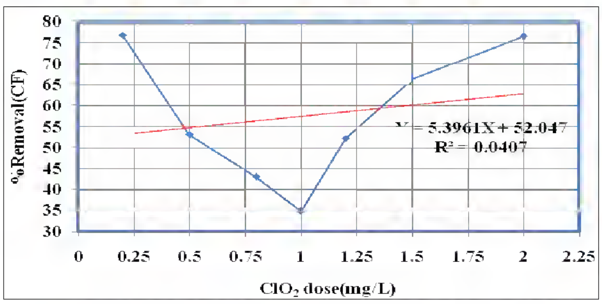


**Figure 6: Percent Reduction in CF with the $ClO_2$ Dose**

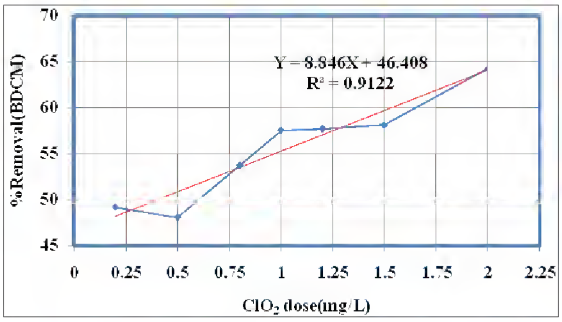


**Figure 7: Percent Reduction in BDCM with the $ClO_2$ Dose**

The correlation is positive between dichlorobromomethane percent reduction and chlorine dioxide dose (decrease in DCBM concentrations with increasing chlorine dioxide dose). Thus, we conclude that DCBM concentration is proportional with $ClO_2$ dose in the form (Y= 8.298x+44.76) with coefficient of determination ($R^2$=0.752) see Figure 8. While positive correlations between bromoform percent reduction and chlorine dioxide dose (slightly decrease in BF concentrations with increasing chlorine dioxide dose). Thus, we conclude that bromoform concentration is proportional with $ClO_2$ dose in the form (Y= 4.927X+49.65) with weak coefficient of determination ($R^2$=0.28) as shown in Figure 9.

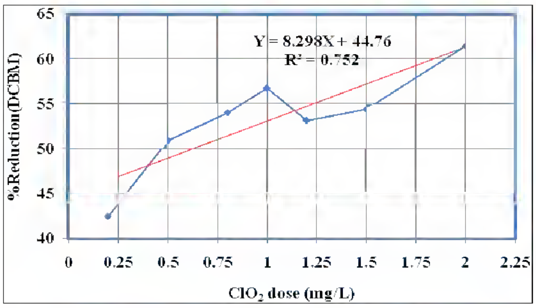


**Figure 8: Percent Reduction in DCBM with the $ClO_2$ Dose**

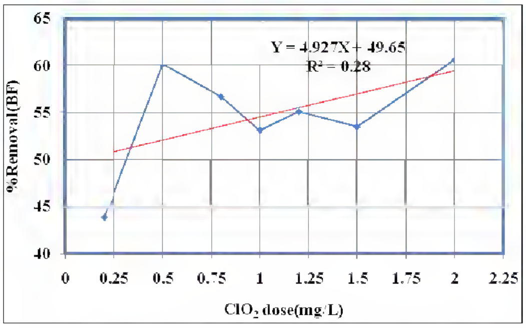


**Figure 9: Percent Reduction in BF with the $ClO_2$ Dose**

## EFFECT OF CONTACT TIME ON TTHMS CONCENTRATION

The effect of detention time onTTHMsformation was investigated with a dosage of 2mg/L chlorine dioxide. The concentration of TTHMs was measured after 30, 60, 90 and120 min. Figure 10 shows that the TTHMs concentration decreases significantly with the increasing contact time. Thus, we conclude that the THMs concentration is proportional with time in the form (Y= -0.0009X+0.1088) with coefficient of determination $R^2$ =0.88. The increasing contact time of $ClO_2$ leads to decrease THMs concentration, the regression equation indicates that, on the average, for 90 min increases in contact time caused decreases of THM4 by about (90.8%).

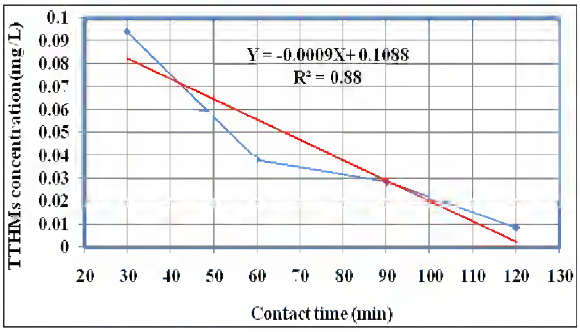


**Figure 10: Variation of TTHMs Concentration with Contact Time**

The variation of individual compound with the contact time is shown in Figure 11. The concentration of chloroform, bromodichloromethane, dibromochloromethane and bromoform decreased with increasing detention time, a production curve almost parallel to thoseTTHMs significantly lower with increasing contact time.

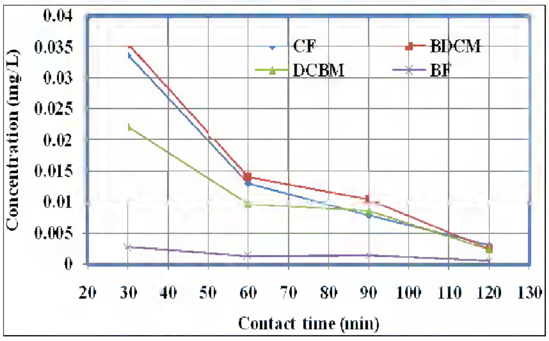


**Figure 11: Variation of TTHMs Compound with Contact Time**

Figure 12 represents the percent reduction in TTHMs concentration along 2 hour detention time. This Figure shows that the largest percent removal of TTHMs is (94.43%) at (120min) of detention time.Thus, we conclude that the reduction in THMs concentration is proportional with time in the form (Y=0.0057X+0.29) with coefficient of determination ($R^2$=0.88).

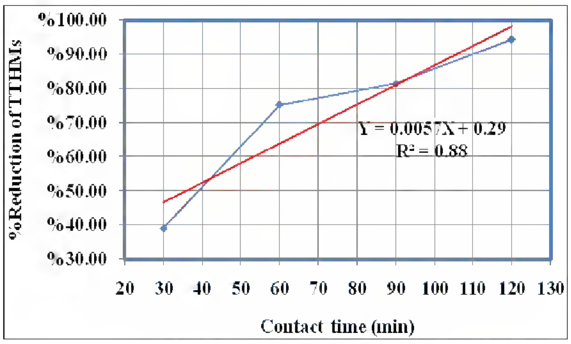


**Figure 12: Percent Reduction in TTHMs with the Contact Time**

When change $ClO_2$ contact time theaverage weight fraction of bromoform compound ranged from 2.97% to 6.4%, while the average weight fraction of $CHBr_3$ was27.72% to 35.8%, for $CHBr_2Cl$ was 30% to 37.65%, and for $CHClBr_2$ was 23.58% to 30.17%) as shown in Figure 13.

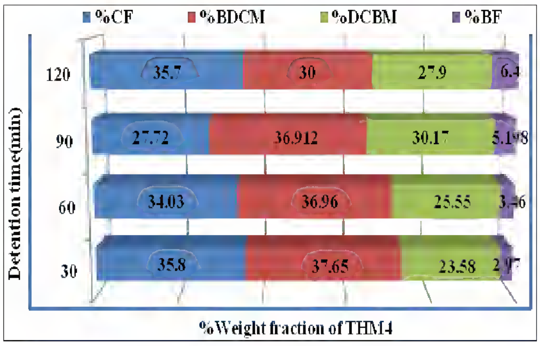


**Figure 13: Weight Fraction of Individual THM4 to Total THM4**

Figure 14 shows the relation between detention time and percent reduction of four compounds (chloroform, bromodichloromethane, dichlorobromomethane, and bromform), as shown the percent removal of four compound of THMs increases with increasing contact time and higher percent removalof THMs compounds is 97.13%, 96%, 95.09%, 87.20% at 120 min for bromoform, dichlorobromomethane, bromodichloromethane, and chloroform respectively.

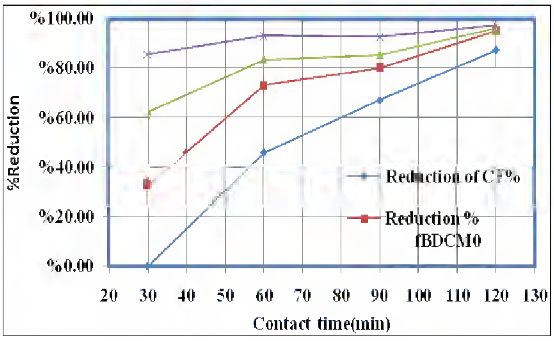


**Figure 14: Percent Reduction of THMs Compounds with the Detention Time**

## TOTAL ORGANIC CARBON CONCENTRATIONS IN TREATED WATER

Figure 15 represents a plot of variations in TOC with various chlorine dioxide doses. As can be seen, the concentrations varied little throughout the entire study period. Additionally, TOC concentrations remaining after each run were not significantly different from one another, but it is decrese with increse $ClO_2$ dose.

Results show that the trend between total organic carbon concentration and chlorine Dioxide Dose began to decreases significantly with the increasing $ClO_2$ dose. Thus, we conclude that the TOC concentration is proportional with $ClO_2$ dose with coefficient of determination ($R^2$ =0.93) in theform(Y=-0.25X +1.438).

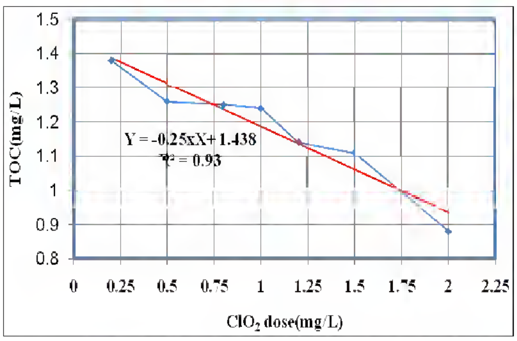


**Figure 15: Variation of Total Organic Carbon with $ClO_2$ Dose**

Figure 16 shows the relation between TTHMs and TOC concentration. The correlations are positive between them; the result shows that the TTHMs concentration increases significantly with the increasing total organic carbon. So, we conclude that the THMs concentration is proportional with TOC in the form (Y=0.046X+0.015) with coefficient of determination ($R^2$ =0.96).

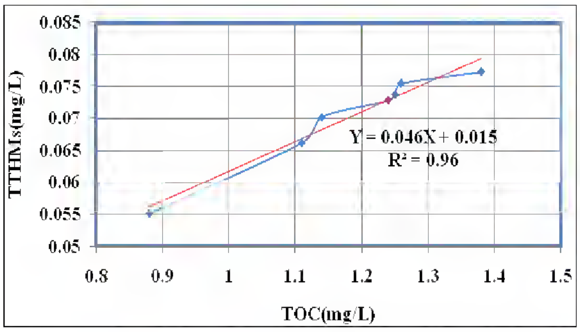


**Figure 16: Variations in TTHMs with TOC**

## EFFECT OF TURBIDITY ON TTHMS FORMATION

Turbidity of filtered water ranges between (4 to7.36) NTU. Lower value when water pre oxidation with (2mg/L) $ClO_2$ dose while the higher value for (0.5mg/L) $ClO_2$.Figure17 shows that increasing in turbidity lead to increase TTHMs concentration, the correlations are positive between them.

Results show that the TTHMs concentration increases significantly with the increasing turbidity. So, we conclude that the THMs concentration is proportional with turbidity, and the correlations are positive between them, in the form (Y= 0.0057X+0.037) with coefficient of determination ($R^2$ =0.85).

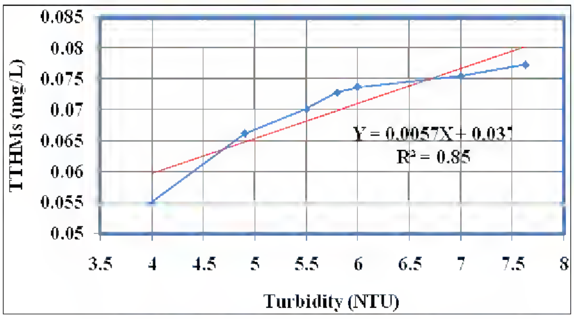


**Figure 17: Variation of Turbidity with Trihalomethane**

Figure 18 shows the variation of four THMs compound with turbidity. As show the result of CF, BDCM, BF, and DCBM variation began slightly decrease with increasing turbidity.

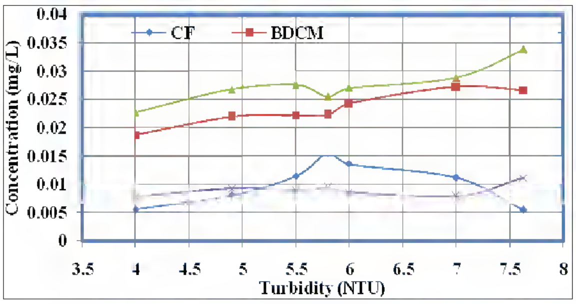


**Figure 18: Variation of THMs Compounds with Turbidity**

## EFFECT OF PH ON TRIHALOMETHANES

Figure19 shows the variation of TTHMs with pH in all runs. It has been found that the degree of inactivation by chlorine dioxide increases as pH increases leads to increases of THMs formation. This finding is agreed with the results adapted by (**Bernarde et al, 1967; Letterman 1999; AWWA, 1982**).

pH effects on THMs formation mechanisms and thus speciation, in which increasing pH lead to increase hydrolysis reactions that many DBPs undergo to form THMs such as trihalopropanones, trihaloacetonitriles, trihaloacetaldes, trihalonitromethanes and trihaloacetic acids(**Nikolaou, 2004).**The relation between pH and THMs formation is in the form (Y=0.018X+0.071) with coefficient of determination $R^2$ equal to (0.973).

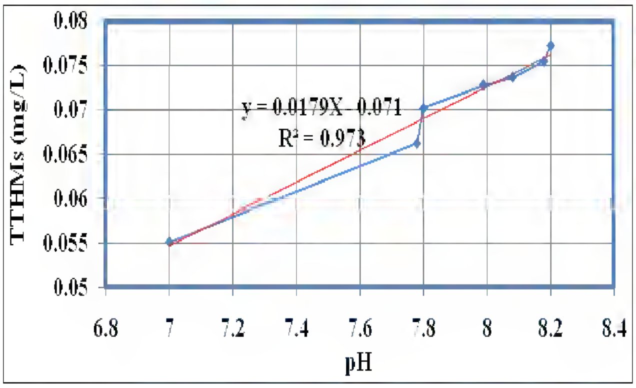


**Figure 19: Variation of pH with TTHMs**

## EFFECT OF TEMPERATURE ON TRIHALOMETHANES

The temperatures ranged in all runes throughout the study period between (24-30) °C. The variation of temperature with THMs is shown in Figure (20). The results show slight variation in TTHMswith temperature, but it is very recognized that TTHMs increase as temperature increase, in the form (Y=0.0032X-0.0154) with coefficient of determination ($R^2$=0.823), because the disinfection efficiency of chlorine dioxide decreases as temperature decreases. This finding is agreed with the results adapted by **(Bernarde, et al., 1967).**

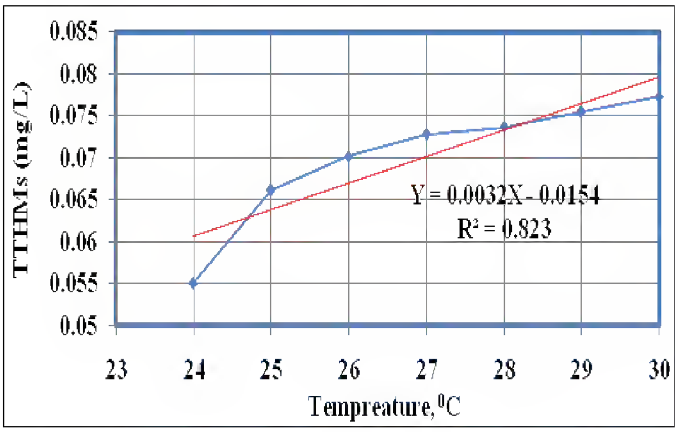


**Figure 20: The Variation of Temperature with TTHM**

## CONCLUSIONS

Based on the objectives of this study the following conclusions and are presented:-

- The results showed that the THM formation decrease with increasing chlorine dioxide dose, concentrations of THMs that developed in water pretreated with 2 mg/L $ClO_2$ were significantly lower when compared to those in water pretreated with the other dose of $ClO_2$.
- The ability of $ClO_2$ to lowering THMs formation was very good. Increasing dose of $ClO_2$ from 1 mg/L to 2 mg/L will caused (24%) reduction of THMs concentrations.
- Chlorine dioxide showed a good potential as an alternative disinfectant and oxidant produced lower THMs concentrations but only at doses higher than (1 mg/L). The optimal dose of $ClO_2$ produced high percent reduction of THM formation (64.33 %) is (2) mg/L.
- The THMs concentration is proportional with time in the form (Y= -0.0009X+0.1088) with coefficient of determination $R^2$ =0.88, increasing of chlorine dioxide contact time from (30 to 120 min) will caused (90.85%) reduction in THMs formation, and higher percent removal of THMs compounds is 97.13% , 96%, 95.09%, 87.20% at 120 min for bromoform, dichlorobromomethane, bromodichloromethane, and chloroform respectively.
- Chloroform concentration is proportional with $ClO_2$ dose in the form (Y= 5.3961X+52.04) with coefficient of determination ($R^2$=0.0407), and the correlation was negative under the dose 1mg/L.
- Correlations are positive between bromodichloromethane and dichlorobromomethane percent reduction and chlorine dioxide dose and slightly decrease in bromoform concentrations with increasing chlorine dioxide dose.
- All results indicate that the treated water contain mainly the brominate THM species ($CHClBr_2$ and $CHBrCl_2$). The average weight fraction of chloroform ($CHCl_3$) compound ranged from 7.16% to 21.46%, while the average weight fraction of the rest brominated ($CHBr_2Cl$, and $CHBrCl_2$) species ranged from about 34.97% to 43.8%, 30.67% to 34.54% respectively.
- Results show that the trend between total organic carbon concentration and chlorine dioxide dose began to decreases significantly with the increasing $ClO_2$ dose, TTHMs concentration increases significantly with the increasing total organic carbon, and turbidity.
- The relation between pH and THMs formation is in the form (Y=0.018X+0.071) with coefficient of determination $R^2$ equal to (0.973), and found that the degree of inactivation by chlorine dioxide increases as pH increases.
- The results show slight variation in TTHMs with temperature, in the form (Y=0.0032X-0.0154) with coefficient of determination ($R^2$=0.823).
- Comparison of total THMs measurements with the USEPA Stage I and stage II limits. Shows that none of the THM levels exceeded the stage I limit, while exceeded the stage II limit in all sampling measurements.
- Dominant TTHMs species during all runs is DCBM, it consists (39.2%) of trihalomethanes.

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